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COHERENTLY PUMPED TWO-FREQUENCY LASER-TYPE DEVICES

Final Report

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I. R. Senitzky

February, 1985

United States Army

EUROPEAN RESEARCH OFFICE OF THE U.S. ARMY

London, England

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A three-level system which radiates at both intermediate transition frequencies is considered to be coherently pumped at a frequency near the two-photon resonance frequency. A sharply resonant rise in the second- and third-level population is found for slight detuning of the pump from two-photon resonance. Population inversion in either transition is shown to be possible, depending on the pertinent parameters. This effect furnishes a novel method of population inversion at a frequency higher than the pump frequency.

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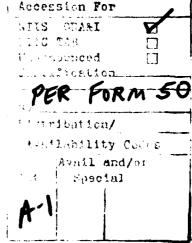
Abstract

A three-level system of intermediate transition frequencies $^{\omega}_{12}$ and $^{\omega}_{23}$, with nonvanishing dipole matrix elements for these transitions only, is considered to be coherently pumped at a frequency near $1/2(^{\omega}_{12} + ^{\omega}_{23})$ and radiating at both intermediate frequencies. A sharply resonant rise in the second—and third-level population is found for slight detuning of the pump from $1/2(^{\omega}_{12} + ^{\omega}_{23})$. Population inversion in either transition is shown to be possible, depending on the pertinent parameters. The induced dipole moment as well as the coherence of the radiation is discussed.

Key Words: Laser, Laser-type Devices, Coherent Pumping,
Two-Photon Pumping, Three-Level Systems, Quantum Electronics







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I. INTRODUCTION

Production of population inversion by means of a "three-level" scheme is one of the standard methods used in lasers and masers pumped by an electromagnetic field. In this scheme, the (1,3) transition and either the (1,2) or the (2,3) transition couple to the electromagnetic field, while the other intermediate transition is nonradiative and must be subjected to a perturbation that produces relaxation. The pump acts on the (1.3) transition, and a steady-state population at the intermediate radiative frequency results for an appropriate range of the pertinent parameters, as follows from conventional rate equations. In order that this scheme be applicable, the states corresponding to the first and third level must be of opposite parity, and the pump frequency must be higher than the laser frequency. In the present report an entirely different method of steady-state population inversion in a three-level system will be described, applicable when the top and bottom states are of the same parity, both intermediate transitions are radiative, and the pump frequency may be either higher or lower than the frequency for which inversion is produced.

II. FORMULATION OF PROBLEM AND DERIVATION OF EQUATIONS OF MOTION

We consider an atomic system ("atom") of three energy levels, $\hbar\omega_1$, $\hbar\omega_2$, $\hbar\omega_3$ in ascending order, with relatively close intermediate transition frequencies ω_{12} , ω_{23} , ($\omega_{ij} \equiv |\omega_i - \omega_j|$) and only two nonvanishing dipole matrix elements $\dot{\mu}_{12}$ and $\dot{\mu}_{23}$, corresponding to the two intermediate transitions, respectively. A pump field E = 250 cosut, with $|\omega_1| = 100$

with resonant frequencies ω_{12} and ω_{23} , respectively, or to the free-space field, described by an infinite, denumerable set of modes. We will refer to the two-mode coupling as Case I, and to the free-space coupling as Case II, distinguishing between these cases, where necessary, by the subscripts I and II, respectively. In the absence of these subscripts, the expression is applicable to either case. For simplicity, we consider the situation in which the field radiated by one transition has a negligible interaction with the dipole moment of the other transition; in Case I, we assume that the ω_{13} mode couples only to the (i,j) transition and in Case II, we assume that different sub-sets of modes couple to the dipole moment of each transition.

In boson second-quantization notation [1], the atomic Hamiltonian is given by $H_a = \Sigma \hbar \omega_1 a_1^{\dagger} a_1$, and the dipole moment by $\vec{d} = \vec{\nu}_{12} a_1 a_2^{\dagger} + \vec{\nu}_{23} a_2 a_3^{\dagger} + h.c.$, where a_1 and a_1^{\dagger} are the usual annihilation and creation operators obeying the commutation rules $[a_1, a_2^{\dagger}] = \delta_{12}$, all other commutators vanishing. Introducing the reduced variables A₂, such that $a_1 = A_2 e^{-i\omega_1 t}$, and using the rotating wave approximation, we express the pump-atom coupling by

$$H_{pa} = H \left[\alpha_{12} A_1 A_2^{+} e^{i(\omega_{12}-\omega)t} + \alpha_{23} A_2 A_3^{+} e^{i(\omega_{23}-\omega)t}\right] + h.c.,$$

where $\alpha_{ij} = -(\vec{E}_0 \cdot \vec{\nu}_{ij})/\hbar$. The field of any mode can be described by the photon annihilation and creation operators b and b⁺, with $(b(t), b^+(t)) = 1$. In Case I, we use the notation bjk, with the corresponding reduced variables given by bjk = Bjk $e^{-i\omega}$ jk^t. In Case II, we use the notation bk, with

 $b_k = B_k e^{-iv} k^t$, where v_k is the frequency of the k'th (free-space) mode. Using, again, the rotating wave approximation [2], we express the coupling between the atom and the field by

where

$$\mathcal{B}_{iji} = Y_{ij} B_{ij}(t)$$

 Y_{ij} being the constant that describes the coupling between the (i,j) transition and the (i,j) cavity mode, and

$$\mathcal{B}_{ijII}(t) = \sum_{k} \gamma_{ijk} B_{K}(t) e^{-i(v_{k} - \omega_{ij})t}$$

 Y_{ijk} being the constant that describes the coupling between the (i,j) transition and the k'th free-space mode. Introducing the notation $\delta \equiv (1/2)\omega_{13}^{-\omega}$ $\Delta \equiv (1/2)(\omega_{12} - \omega_{23})$, one obtains, as atomic equations of motion

$$\dot{A}_{1} = -i(\alpha_{12}A_{2}e^{-i(\delta+\Delta)t} + \mathcal{B}_{12}^{+}A_{2}) ,$$

$$\dot{A}_{2} = -i(\alpha_{12}A_{1}e^{i(\delta+\Delta)t} + \alpha_{23}A_{3}e^{-i(\delta-\Delta)t} + A_{1}\mathcal{B}_{12} + \mathcal{B}_{23}^{+}A_{3}) ,$$

$$\dot{A}_{3} = -i(\alpha_{23}A_{2}e^{i(\delta-\Delta)t} + A_{2}\mathcal{B}_{23}) .$$

Expression for the field in terms of atomic variables are obtained from previously derived results. These are [3]

$$\mathcal{B}_{ijI}(t) = \gamma_{ij} B_{ij}(0) - i |\gamma_{ij}|^2 \int_0^t dt_1 A_i^+(t_1) A_j(t_1) e^{-\xi_{ij}(t-t_1)}, i < j$$

where ξ_{1} is the loss constant of the (i,j) mode (energy will decay freely as exp (-2 ξ_{1}), and [4]

$$\mathcal{B}_{ijII}^{(t)} = \mathcal{B}_{ijII}^{(0)}(t) - ik_{ij} A_{i}^{+}(t)A_{j}(t), i < j$$

where k_ij is a positive constant proportional to the average value (averaged over the index k) of $|Y_{ijk}|^2$ in the neighborhood of $v_k = \omega_{ij}$, and $\beta_{ij}^{(0)}$ is the unperturbed β_{ij} , that is, the operator obtained by replacing $B_k(t)$ by $B_k(0)$ in the expression for $\beta_{ijjj}(t)$. (It is assumed that the pump is turned on at t=0.) We will consider, presently, only the situation in which the loss in the cavity-modes is sufficiently large so that the field follows the atomic polarization adiabatically; in other words, ξ_{ij} is larger than the rate of change of $\langle A_i^{\dagger}A_j \rangle$. In this situation one can write, for $t \rangle \xi_{ij}^{-1}$,

$$\mathcal{B}_{ijI}^{(t)} = \mathcal{B}_{ijI}^{(0)} - i k_{ijI} A_{i}^{+}(t) A_{j}^{(t)},$$
where
$$k_{ijI} = |\gamma_{ij}|^{2} / \xi_{ij}, \quad \mathcal{B}_{ijI}^{(0)} = \gamma_{ij} B_{ij}^{(0)}.$$

Since $\mathcal{B}_{ij}^{(0)}$ i) = 0 for both Case I and Case II, the expression for $\mathcal{B}_{ij}(t)$ has the same form in both cases. It is therefore unnecessary to treat them separately, and the subscripts I and II will be dropped with the understanding that the discussion, henceforth refers to both cases.

Our present interest lies in the steady-state atomic level populations and dipole moments. We need, therefore, equations for the expectation values of all bilinear products $\Lambda_1^{\dagger}\Lambda_1$ with respect to an (initially) unexcited radiation field. (It should be noted that the quantities $\langle \Lambda_1^{\dagger}\Lambda_1 \rangle$ are the elements ρ_{ij} of the atomic density matrix in the interaction representation.) The

derivation of such equations from those for the Ai's in a convenient form is outlined briefly in the following.

We first write expressions for all derivatives $(d/dt) < h_1^{\dagger} h_1$ and order each term so that B_{11}^{\dagger} appears on the extreme left and B_{11}^{\dagger} appears on the extreme right. For the case of an initially unexcited radiation field, the B_{11}^{\dagger} term refers to the vacuum field, and the k_{11} term accounts for the radiation reaction. Substituting for each $B_{11}(t)$ these two terms, and utilizing the relationship $<|B_{11}^{\dagger}|^2 + (t) = B_{11}^{\dagger}|^2 + (t) = 0$, we drop all explicit vacuum-field references. The above ordering of the 11's also determines the ordering of the four atomic factors in the k_{11} terms, which we now rewrite in normal order, using the appropriate commutation relationships. Utilizing another previously derived result [1], namely,

 A_1A_2 !> = 0 when the state !> describes a single atom, we drop all four-factor terms (remaining after the normal ordering). Finally, the substitutions

$$A_1 = x_1 e^{-i\delta t}$$
, $A_2 = x_2 e^{i\delta t}$, $A_3 = x_3 e^{i\delta t}$,

$$P_{ij} = (x_i x_j^+ + x_i^+ x_j^-), S_{ij} = i(x_i x_j^+ - x_i^+ x_j^-),$$

are used to obtain equations of motion with constant coefficients for nine real variables:

$$\begin{split} &\mathring{\mathbf{n}}_{1} = \alpha_{12} \mathbf{S}_{12} + 2\mathbf{k}_{12} \mathbf{n}_{2} \\ &\mathring{\mathbf{n}}_{2} = -\alpha_{12} \mathbf{S}_{12} + \alpha_{23} \mathbf{S}_{23} - 2\mathbf{k}_{12} \mathbf{n}_{2} + 2\mathbf{k}_{23} \mathbf{n}_{3} \\ &\mathring{\mathbf{n}}_{3} = -\alpha_{23} \mathbf{S}_{23} - 2\mathbf{k}_{23} \mathbf{n}_{3} \\ &\mathring{\mathbf{s}}_{12} = -(\delta + \Delta) \mathbf{P}_{12} - 2\alpha_{12} (\mathbf{n}_{1} - \mathbf{n}_{2}) - \alpha_{23} \mathbf{P}_{13} - \mathbf{k}_{12} \mathbf{S}_{12} \\ &\mathring{\mathbf{s}}_{23} = -(\delta - \Delta) \mathbf{P}_{23} - 2\alpha_{23} (\mathbf{n}_{2} - \mathbf{n}_{3}) + \alpha_{12} \mathbf{P}_{13} - (\mathbf{k}_{12} + \mathbf{k}_{23}) \mathbf{S}_{23} \\ &\mathring{\mathbf{s}}_{13} = -2\delta \mathbf{P}_{13} - \alpha_{23} \mathbf{P}_{12} + \alpha_{12} \mathbf{P}_{23} - \mathbf{k}_{23} \mathbf{S}_{13} \\ &\mathring{\mathbf{p}}_{12} = (\delta + \Delta) \mathbf{S}_{12} + \alpha_{23} \mathbf{S}_{13} - \mathbf{k}_{12} \mathbf{P}_{12} \\ &\mathring{\mathbf{p}}_{23} = (\delta - \Delta) \mathbf{S}_{23} - \alpha_{12} \mathbf{S}_{13} - (\mathbf{k}_{12} + \mathbf{k}_{23}) \mathbf{P}_{23} \\ &\mathring{\mathbf{p}}_{13} = 2\delta \mathbf{S}_{13} + \alpha_{23} \mathbf{S}_{12} - \alpha_{12} \mathbf{S}_{23} - \mathbf{k}_{23} \mathbf{P}_{13} \\ &\mathring{\mathbf{p}}_{13} = 2\delta \mathbf{S}_{13} + \alpha_{23} \mathbf{S}_{12} - \alpha_{12} \mathbf{S}_{23} - \mathbf{k}_{23} \mathbf{P}_{13} \\ &\mathring{\mathbf{p}}_{13} = 2\delta \mathbf{S}_{13} + \alpha_{23} \mathbf{S}_{12} - \alpha_{12} \mathbf{S}_{23} - \mathbf{k}_{23} \mathbf{P}_{13} \\ &\mathring{\mathbf{p}}_{13} = 2\delta \mathbf{S}_{13} + \alpha_{23} \mathbf{S}_{12} - \alpha_{12} \mathbf{S}_{23} - \mathbf{k}_{23} \mathbf{P}_{13} \\ &\mathring{\mathbf{p}}_{13} = 2\delta \mathbf{S}_{13} + \alpha_{23} \mathbf{S}_{12} - \alpha_{12} \mathbf{S}_{23} - \mathbf{k}_{23} \mathbf{P}_{13} \\ &\mathring{\mathbf{p}}_{13} = 2\delta \mathbf{S}_{13} + \alpha_{23} \mathbf{S}_{12} - \alpha_{12} \mathbf{S}_{23} - \mathbf{k}_{23} \mathbf{P}_{13} \\ &\mathring{\mathbf{p}}_{13} = 2\delta \mathbf{S}_{13} + \alpha_{23} \mathbf{S}_{12} - \alpha_{12} \mathbf{S}_{23} - \mathbf{k}_{23} \mathbf{P}_{13} \\ &\mathring{\mathbf{p}}_{13} = 2\delta \mathbf{S}_{13} + \alpha_{23} \mathbf{S}_{12} - \alpha_{12} \mathbf{S}_{23} - \mathbf{k}_{23} \mathbf{P}_{13} \\ &\mathring{\mathbf{p}}_{13} = 2\delta \mathbf{S}_{13} + \alpha_{23} \mathbf{S}_{12} - \alpha_{12} \mathbf{S}_{23} - \mathbf{k}_{23} \mathbf{P}_{13} \\ &\mathring{\mathbf{p}}_{13} = 2\delta \mathbf{S}_{13} + \alpha_{23} \mathbf{S}_{13} - \alpha_{12} \mathbf{S}_{13} - \mathbf{k}_{23} \mathbf{P}_{13} \\ &\mathring{\mathbf{p}}_{13} + \alpha_{23} \mathbf{S}_{13} - \mathbf{k}_{23} \mathbf{S}_{13} \\ &\mathring{\mathbf{p}}_{13} + \alpha_{23} \mathbf{S}_{13} - \alpha_{12} \mathbf{S}_{13} - \mathbf{k}_{23} \mathbf{S}_{13} \\ &\mathring{\mathbf{p}}_{13} + \alpha_{23} \mathbf{S}_{13} - \alpha_{12} \mathbf{S}_{13} - \mathbf{k}_{23} \mathbf{S}_{13} \\ &\mathring{\mathbf{p}}_{13} + \alpha_{23} \mathbf{S}_{13} - \alpha_{12} \mathbf{S}_{13} - \mathbf{k}_{23} \mathbf{S}_{13} \\ &\mathring{\mathbf{p}}_{13} + \alpha_{23} \mathbf{S}_{13} - \alpha_{12} \mathbf{S}_{13} - \mathbf{k}_{23} \mathbf{S}_{13} \\ &\mathring{\mathbf{p}}_{13} + \alpha_{12} \mathbf{S}_{13} - \alpha_{12} \mathbf{S}_{13} - \mathbf{k}_{23} \mathbf{$$

These equations exhibit $2k_{13}$ and $2a_{13}$ as the decay rate and Rabi frequency, respectively, associated with the (i,j) transition. When multiplied by the appropriate matrix elements, P_{12} and P_{23} describe the components of the dipole-moment expectation values in phase with the field, while S_{12} and S_{23} describe those in quadrature with the field (which are responsible for the power absorption). It should be noted that these expectation values represent only the coherent part of the dipole moment. The incoherent part must be obtained from other than the lowest moments. The coherence of the radiation will be discussed later.

III. SOLUTION OF EQUATIONS OF MOTION

As mentioned previously, our present interest lies in a steady-state solution. Setting the time derivatives equal to zero, and replacing one of the first three equations, (say, that for n_2) by $[n_i = 1]$, we obtain a set of nine inhomogeneous

equations for nine unknowns. An exact analytic solution is formally possible but complicated, and we resort at first to a numerical solution. For this purpose all the constants, which are inverses of the time, can be normalized by taking a12 to be unity; all frequencies and decay constants are then given in terms of the (1,2) Rabi frequency. Figure 1 shows a three-dimensional graph of n_2/n_1 vs. δ and Δ . The resonance that appears in n_2/n_1 for large Δ is striking, and motivates an approximation that makes analytic solutions of the steady-state equations much simpler. We consider the case $\Delta >> \alpha_{ij} >> k_{ij}$. If the time scale is chosen so that $\alpha_{ij}-1$, one can regard Δ^{-1} and k_ij as small quantities (of first order) compared to unity, and examine the solution for small δ . The four equations containing Δ can now be approximated by retaining only the lowest order terms, that is, by dropping the terms (in these equations only) containing & and k. The resulting set of steady state equations yields a solution which can be written most succintly as follows: 1.et

$$r \equiv k_{12}/k_{23}$$
, $\delta_0 \equiv (a_{23}^2 - a_{12}^2)/2\Delta$,

and

$$R = \frac{\Delta^2}{\alpha_{12}^2 \alpha_{23}^2} [k_{23}^2 + 4(\delta - \delta_0)^2]$$

With this notation, we have

$$n_{2} = [1+r(2+R)]^{-1},$$

$$n_{1} = r(1+R)n_{2},$$

$$n_{3} = rn_{2},$$

$$s_{12} = -2(k_{12}/\alpha_{12})n_{2},$$

$$s_{23} = -2(k_{12}/\alpha_{23})n_{2},$$

$$s_{13} = 2(\Delta k_{12}/\alpha_{12}\alpha_{23})n_{2},$$

$$P_{12} = 2\left\{\frac{\alpha_{12}}{\Delta}(1-2) - \frac{\Delta k_{12}k_{23}}{\alpha_{12}\alpha_{23}}^{2} - \frac{2(\delta-\delta_{0})r}{\alpha_{12}}\left[1 + \frac{2\Delta(\delta-\delta_{0})}{\alpha_{23}^{2}}\right]\right\} n_{2}$$

$$P_{23} = 2\left[\frac{\alpha_{23}}{\Delta}(1-r) - 2r\frac{(\delta-\delta_{0})}{\alpha_{23}}\right] n_{2},$$

$$P_{13} = 4r\frac{\Delta(\delta-\delta_{0})}{\alpha_{12}\alpha_{23}} n_{2}.$$

Figure 2 displays both the numerical solution of the exact equations and the analytic solution of the approximate equations for the level-populations as functions of $^{\delta}$. It is seen that, for the parameters used, the approximation is good.

IV. DISCUSSION OF SOLUTIONS

Since the quantity R reaches a minimum at $\delta = \delta_0$, the analytic solutions for n_2 and n_3 , as well as those for $|S_{12}|$, $|S_{23}|$ and $|S_{13}|$, all exhibit resonant-type maxima as a function of pump frequency at (1/2) ω_{13} - δ_0 , that is, when the pump is detuned from exact two-photon resonance by δ_0 . The half-maximum resonance width about δ_0 for either n_2 or n_3 is given by

$$2(\delta-\delta_0)_{\frac{1}{2}\max} = \left[\frac{\alpha_{12}^2\alpha_{23}^2}{\Delta^2} \left(2 + \frac{k_{23}}{k_{12}}\right) + k_{23}^2\right]^{\frac{1}{2}} ,$$

and that for n2/n1 is given by

$$2(\delta-\delta_0)_{\frac{1}{2}\max} = \left(\frac{\frac{2}{\alpha_{12}\alpha_{23}^2}^2}{\frac{2}{\Delta^2}} + k_{23}^2\right)^{\frac{1}{2}}$$

$$(n_2/n_1)_{\text{max}} = r^{-1} (1 + \Delta^2 k_{23}^2 \alpha_{12}^{-2} \alpha_{23}^{-2})^{-1}$$

The transition at which inversion occurs will have either a higher or a lower frequency than the pump frequency, the difference being approximately Δ . It is also interesting to note that P13 goes through zero at resonance.

It is clear that the present effect may have interesting applications, firstly, because population inversion for a given pair of levels may not be otherwise achievable, secondly, because inversion can be achieved at a higher frequency than the pump frequency, and thirdly, because a sharp resonance has many obvious uses in physics. Discussion of the details of such applications is beyond the scope of the present paper.

V. RATE-PROCESSES ANALYSIS

It is desirable to offer a more intuitive type of explanation of the present resonance effect than that contained in the mathematics. The first part of such an explanation is the observation that $(1/2)^{\omega_1}3^{-\delta_0}$ is just the pump frequency at which a three-level system such as the present, but without radiation losses (that is, without coupling to the radiation field), behaves like a two-level system for \$ >> aij[5]. Its population, if intially in the ground state, mainly oscillates between the first and the third level (a Rabi-type oscillation) with frequency $2a_{12}a_{23}/\Delta$, while the second-level population remains small $(-\alpha_{12}^2/\Delta^2)$ and oscillates with relatively high frequency $(-\Delta)$ [6]. We can regard our present radiative system as responding to the pump like this lossless system, pumped from level 1 to level 3 by a two-photon process. The second part of the explanation is the observation that the present system behaves like a three-level system with respect to radiation losses, relaxing at both intermediate frequencies with a consequent distribution of population to all levels.

Since the above explanation invokes, implicity, rate processes, it is of interest to see how it can be presented in quantitative form using rate equations. (It should be noted that the explanation is applicable only to the range of parameter values for which the approximation was made.) A set of rate equations for three levels reads,

 $\dot{n}_1 = R_{12}(n_2-n_1) + R_{13}(n_3-n_1) + 2k_{12}n_2$

 $n_2 = R_{12}(n_1-n_2) + R_{23}(n_3-n_2) + 2k_{23}n_3-2k_{12}n_2$

 $n_3 = R_{23}(n_2-n_3) + R_{13}(n_1-n_3) - 2k_{23}n_3$

where R_{iJ} is the pumping rate between levels i and J. In order to obtain the pumping rate between consecutive levels, we look at the equations of motion for a two-level system. These can be obtained immediately from the three-level equations of motion by eliminating all references to either the first or the third level. Dropping all references to the third level, we obtain

$$\dot{n}_1 = \alpha_{12} + 2k_{12} + \alpha_{12}, \qquad \dot{n}_2 = -\dot{n}_1,$$

$$\dot{s}_{12} = - (\delta + \Delta)P_{12} - 2\alpha_{12}(n_1 - n_2) - k_{12}S_{12}$$

$$\dot{P}_{12} = (\delta + \Delta)S_{12} - k_{12}P_{12}$$

where the deviation of the pump frequency from resonance is now given by $\delta+\Delta$. Since the rate equations for two levels reduce to

 $\dot{n}_1 = R_{12}(n_2-n_1) + 2k_{12}n_2, \quad \dot{n}_2 = -\dot{n}_1,$

we must have

 $R_{12}(n_2-n_1) = {}^{\alpha}_{12}5_{12},$

Implicit in the use of rate equations is the assumption that rates are independent of the state of the system. We consider the steady state, set $\dot{S}_{12} = \dot{P}_{12} = 0$, eliminate P_{12} from the last two equations of motion, and obtain

$$a_{12}S_{12} = \frac{2a_{12}^2k_{12}}{k_{12}^2 + (\delta + \Delta)^2} (n_2 - n_1),$$

which yields

$$R_{12} = \frac{2\alpha_{12}^2 k_{12}}{k_{12}^2 + (\delta + \Delta)^2} ,$$

a generally familiar result.

We return, now, to the three-level rate equation and compare them with the three-level equations of motion, obtaining thus $\alpha_{12}S_{12} = R_{12}(n_2-n_1) + R_{13}(n_3-n_1).$

For the range of parameters that led to the approximate equations of motion, R_{12} is a small quantity of the order of k_{12}/Δ^2 , that is, of third order. (It should be recalled that k_{13} and Δ^{-1} are small quantities of first order, and α_{13} is of order unity.) From the steady state equations, we see that $\alpha_{12}S_{12}$ must be of order k_{12} , since it equals $-2k_{12}n_2$, and n_1 is, of course, of order unity. R_{12} is therefore negligible compared to R_{13} , and R_{13} must be of the same order as S_{12} . A similar argument applies to R_{23} relative to R_{13} . One can say that the approximations made in the equations of motion find their counterpart, in the rate equations, in the neglect of R_{12} and R_{23} compared to R_{13} . Writing the three-level rate equations in accordance with this prescription we have

n1 = R13(n3-n1) + 2k12n2,

n₂ ≈ 2k₂3n₃ - 2k₁2n₂,

ng ≈ R13(n1-n3) - 2k23n3.

This is just the quantitative form of the qualitative explanation offered earlier. If we now set

$$R_{13} = \frac{2\alpha_{12}^2 \alpha_{23}^2 k_{23}}{\Delta^2 k_{23}^2 + 4(\delta - \delta_0)^2} ,$$

these rate equations will yield exactly the same expression for the steady-state populations as the analytic solution of the full (approximate) equations of motion. It should be noted that,

while rates for rate-equation purposes are usually taken to be the transition probabilities derived by perturbation theory, the above expression for R₁₃, with the detuning, can come only from consideration of the full set of coherent equations. Lastly, as a check, we compare the rate of energy absorption from the pump with the rate of energy loss to the field. The former is given by

 $A(\omega_{12} + \omega_{23})R_{13}(n_1-n_3)$ while the latter is given by

2ή ω_{12k12n2} + 2ήω_{23k23n3} .

Substitution of the above expression for R_{13} and the analytic solutions for n_1 in these two rates exhibits their equality. VI. COHERENCE

It is of interest to examine the coherence properties of the radiation emitted by the pumped atom. For simplicity, we use the notation applicable to Case I, in which the atom radiates into two lossy cavity modes. The (complex) field operator for the (ij) mode is proportional to Bij $\exp(-i\omega_1)t$). By factoring out γ_{ij} from the expression for ijI(t) in Sec. II, we obtain

$$B_{ij}(t) = B_{ij}(0) - \frac{i\gamma_{j}^{*}}{\xi_{ij}} A_{i}^{+}(t) A_{j}(t)$$
, i < j ,

where, it is recalled, $B_{ij}(0)! > = 0$. The coherent field is proportional to $\langle B_{ij}(t) \rangle \exp(-i\omega_{ij}t)$. The expectation value of the total energy in the (i,j) mode is $\Delta \omega_{ij} \langle B_{ij}^{\dagger} B_{ij} \rangle$, while the energy associated with the coherent field is $\Delta \omega_{ij} \langle B_{ij} \rangle \langle B_{ij} \rangle$. From the above operator expression for $\Delta \omega_{ij} \langle B_{ij} \rangle \langle B_{ij} \rangle$.

$$\langle B_{1j}^{+}B_{1j}^{-}\rangle = \frac{|\gamma_{1j}|^{2}}{\xi_{1j}^{2}} \langle A_{1}A_{j}^{+}A_{1}^{+}A_{j}^{-}\rangle = \frac{k_{1j}}{\xi_{1j}} n_{j}, \quad 1 < j,$$

(since A_1A_310) = 0 for a single atom), and

$$\langle B_{ij}^{+} \rangle \langle B_{ij} \rangle = \frac{1}{4} (k_{ij}/\xi_{ij}) (P_{ij}^{2} + S_{ij}^{2})$$
,

where (i,j) stands for either (1,2) or (3,4). The ratio of the coherent field energy to the total energy is, therefore, given by

$$\frac{\langle B_{ij}^{+} \rangle \langle B_{ij}^{+} \rangle}{\langle B_{ij}^{+} B_{ij}^{+} \rangle} = \frac{{}^{1}_{4}(P_{ij}^{2} + S_{ij}^{2})}{{}^{n}_{j}}.$$

The order of magnitude of this quantity is the same as that of $(k_1J/\alpha_1J)^2 + (\alpha_1J/\Delta)^2$, which is a small quantity of second order for the parameter range under consideration. We see, therefore, that most of the radiation is incoherent. The frequency of the coherent radiation is that of the pump, as is to be expected; the incoherent radiation, on the other hand, may be expected to lie, largely, in the frequency regions near the two transition frequencies. A quantitative discussion of the spectral distribution of the radiation is beyond the scope of the present paper.

VII. CONCLUSION AND SUMMARY

Single-frequency two-photon pumping of a three-level system that radiates naturally only at the two intermediate frequencies has been considered. It has been shown that when the pump is slightly detuned from two-photon resonance and not too close to

one-photon resonance for one of the intermediate frequencies, population inversion can be obtained either in the upper or in the lower pair of levels, depending mainly on the ratio of the two radiative decay constants. This effect furnishes a novel method of population inversion for laser purposes and a offers the possibility of obtaining population inversion in a transition with a frequency higher than the pump frequency.

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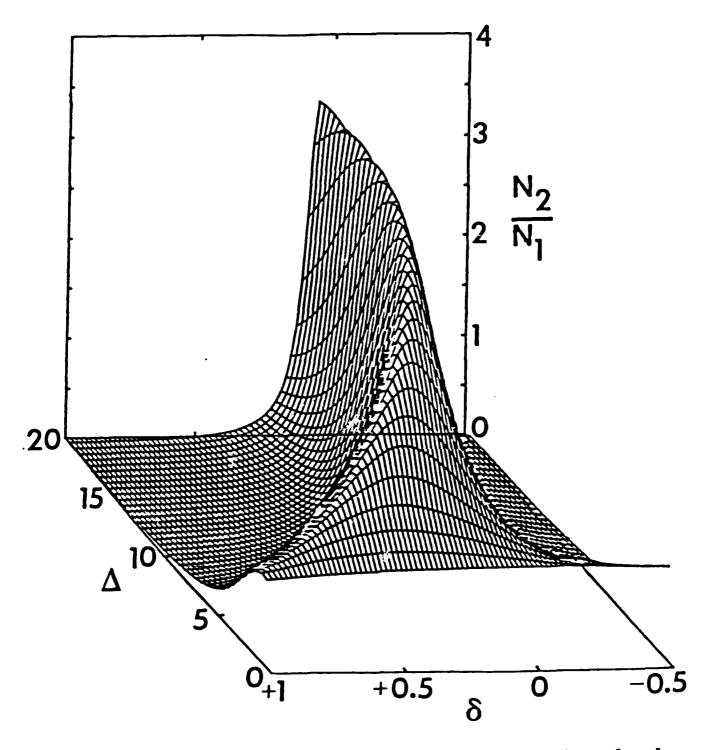


Fig. 1 The steady state population ratio n_2/n_1 in a three-level system as a function of $\delta = \frac{1}{2}\omega_{13} - \omega$ and $\Delta = \frac{1}{2}(\omega_{12} - \omega_{23})$ for $\alpha_{12} = 1, \ \alpha_{23} = 2, \ k_{12} = 0.01 \text{ and } k_{23} = 0.04. \text{ (We choose } (k_{23}/k_{12}) = (\alpha_{23}/\alpha_{12})^2, \text{ since } \alpha_{ij} = (\frac{1}{\nu}_{ij})^2 \text{ and } k_{ij} = (\frac{1}{\nu}_{ij})^2.)$

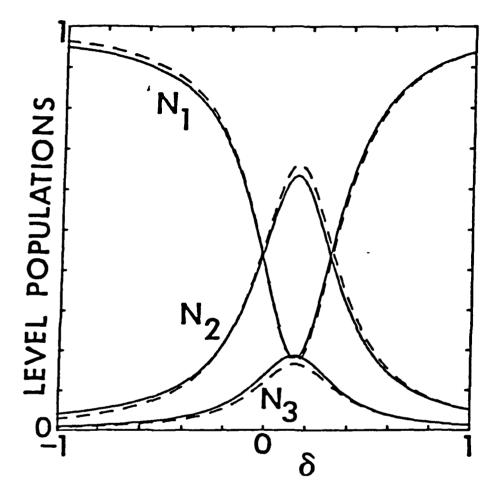


Fig. 2 A comparison of the exact numerical solution (solid lines) and approximate analytic expressions (dashed lines) for the steady state populations as functions of δ , with $\alpha_{12} = 1$, $\alpha_{23} = 2$, $k_{12} = 0.01$, $k_{23} = 0.04$ and $\Delta = 10$.

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